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PATENT
Attorney Docket No. 628365009012

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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In re Application of	:	Hayakawa, et al.
Serial No.	:	09/374,344
Filed	:	August 13, 1999
For		Method for Photocatalytically Rendering a Surface of a Substrate Superhydrophilic, a Substrate with Superhydrophilic Photocatalytic Surface, and Method of Making Thereof
Art Unit	:	1762
Examiner	:	Padgett, M.

SUBMISSION OF DECLARATION OF TOSHIYA WATANABE

Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

Sir:

Submitted herewith is the Declaration of Toshiya Watanabe pursuant to 37 C.F.R. 1.132. This Declaration is cited in the Preliminary Amendment and Supplemental Response filed on July 24, 2003.

Respectfully submitted

Calvin P. Griffith, Registration No. 34,831
JONES DAY
North Point, 901 Lakeside Avenue
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Attorney for Toto Limited

Date: August 1, 2003

I hereby certify that this document is being faxed to the USPTO this 1st Day of August, 2003

Pamela J. Clyngier

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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Applicants	:	Hayakawa, et al.
Application No.	:	09/374,344
Title	:	Method for Photocatalytically Rendering a Surface of a Substrate Superhydrophilic, a Substrate with a Superhydrophilic Photocatalytic Surface, and Method of Making Thereof
Filing Date	:	August 13, 1999
Examiner	:	M. Padgett
Group Art Unit	:	1762
Attorney Docket	:	628365009012

DECLARATION OF TOSHIYA WATANABE
PURSUANT TO 37 C.F.R. 1.132

I, Toshiya Watanabe, declare and state as follows:

1. I am currently employed at the University of Tokyo as Professor in the Research Center of Advanced Science and Technology. Until 1999 I was employed at Toto Ltd. ("Toto"). My last position at Toto was Executive Assistant Researcher in Toto's Basic Research Center. In that and earlier positions, I had responsibility for research and development in the area of superhydrophilic photocatalytic coatings. I am one of the named inventors on the above-referenced patent application, as well as U.S. Patent No. 6,013,372. I am also a named inventor on other patents relating to superhydrophilic photocatalytic coatings.

2. In 1982 I received an Engineering Bachelor's degree in Inorganic Material Engineering. In 1984 I received an Engineering Master's degree in Inorganic Material Engineering. In 1994 I received an Engineering Doctorate in Inorganic Material Engineering. All of these degrees were received from the Tokyo Institute of Technology.

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3. I am a co-inventor on numerous patents in the areas of dielectric ceramics, electrostatic devices, discharge devices, photocatalysts, ozone oxidation processes, and sol-gel coatings. I have also authored or co-authored numerous original papers and review articles. I am an author or co-author of four books, including one in English. The book in English is:

A. Fujishima, K. Hashimoto and T. Watanabe
TiO_2 Photocatalysis Fundamentals and Applications
Bkc, Inc. (1999)

I have also given presentations at either professional society meetings or academic, industrial or governmental research centers in Japan, the United States, Canada, China, Germany and Italy.

4. In 1998 I received the Innovation in Raw Materials award for my work on photocatalytic coatings. In 1999 I received the D.R. ULRICH Award for my presentation at 10th International Workship on Glass, Ceramics, Hybrids, and Nanocomposites from Gels.

5. I have reviewed Japanese Laid-Open Patent Application No. 149281/1978 to Okaniwa et al. ("Okaniwa"). That application describes making a hydrophilic film laminate. The hydrophilic film laminate is comprised of: (1) a substrate; and (2) a film provided on the substrate, the film having been hydrophilified by irradiating a hydrolyzate of a tetraalkoxide of titanium with ultraviolet light.

6. In Table 1 Okaniwa provides certain details of the conditions under which the film is formed. Okaniwa describes using either a #5 or #10 bar coater to coat a film of "TBT" to a thickness of about 300 angstroms. "TBT" is described as being a "monomer, a tetramer, and a decamer of tetra-n-titanate." From the table, it appears that each coating was either a monomer, or a tetramer, or a decamer, rather than being a mixture of the three. The coatings were air-dried and, in some cases, heated to a temperature of 110°C for 3 minutes.

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7. In Table 2 certain other examples are set forth. In these examples, Okaniwa describes making "TPT" films from a monomer of tetraisopropyl titanate. The coatings were air-dried but, apparently, not heated (since no conditions are reported for heating the coatings in Table 2).

8. Okaniwa describes that ultraviolet irradiation was applied to the coatings described in the examples. The irradiation times were either 1, 3 or 5 minutes. Okaniwa reports that after UV irradiation, the contact angle with water decreased. The longer the time period for irradiation, the greater the decrease in water contact angle.

9. In my opinion, the amount of heating of the alkoxide of titanium in the Okaniwa examples would be insufficient to generate appreciable amounts of crystalline titania. In our patent application (page 36, Table 1), we describe heating conditions of alkoxides of titanium to much higher temperatures, and we found that only at 475°C or higher were anatase crystals confirmed. At 450°C we did not confirm the presence of anatase crystals. We also found that this had an impact on the degree of decrease of water contact angle after UV irradiation. Further, the titania species formed at lower temperatures did not appear to be operating as photocatalysts. Therefore, the species do not show photo-induced hydrophilicity.

10. In the Okaniwa examples the alkoxide of titanium coatings are heated to a temperature of much less than 450°C. The highest heating level reported in Okaniwa is 110°C. As a result, in my opinion, any decrease in water contact angle in the Okaniwa examples is not due to photo-induced hydrophilic action of titania, but rather, from other causes, such as increasing the amount of hydrophilic chemical species present in the coating. On page 2 of Okaniwa there is a description of various hydrolyzates of the alkoxides of titanium, and, as the reaction is driven to the right in this series of reactions, the hydrophilicity of the various species

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tends to increase. UV irradiation may help cause these reactions to be driven to the right, thus increasing hydrophilicity without the use of photocatalysis or a photocatalyst. In fact, Okaniwa does not mention photocatalysis and does not describe any of its components as being photocatalysts.

11. I declare under penalty of perjury that the foregoing is true and correct.

July 25, 2003

Toshiya Watanabe

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